# PHYTOCHEMICAL ASPECTS OF PHYLOGENY IN HAMAMELIDAE[1]

DAVID E. GIANNASI[2]

ABSTRACT

Chemical data for the Hamamelidae (sensu Cronquist) are numerous but scattered. Few large-scale comprehensive surveys of any particular group of compounds (micro- or macromolecular) exist for the Hamamelidae. This has limited the use of such data in drawing broad systematic conclusions beyond those based on extant morphological, anatomical, and palynological studies. Certainly, available data for some classes of compounds, such as phenolics, have proven useful at the inter- and intrafamilial level (e.g., Ulmaceae, Juglandaceae, Urticaceae). However, the diverse and sometimes "exotic" micromolecules (e.g., alkaloids, sesquiterpene lactones, polyacetylenes, glucosinolates) often found in the other subclasses of angiosperms are mostly lacking in the Hamamelidae. This implies, at least from present surveys, a biochemical conservatism (or alternatively, reduction) for the group and an early and considerable divergence from its more chemically diverse putative Magnoliid ancestors.

In an earlier review of the phytochemistry of the "Amentiferae," Mears (1973) catalogued the various classes of secondary metabolites for the group, including phenolics, sugars, various types of terpenoids (including iridoids), several alkaloids, and fatty acids. His major conclusions were that (1) insufficient comprehensive surveys of any class of secondary metabolites were available and thus (2) few correlations or putative relationships between taxa in the "Amentiferae" could be drawn.

More than ten years later (using computer-assisted and manual literature surveys), basically the same conclusions may be drawn despite a moderate increase in the number of new compounds discovered and an equally moderate increase in the number of families surveyed in detail for any single class of compounds (mostly phenolics). This is surprising for a group that has undergone considerable taxonomic redefinition and reemerged as the Hamamelidae (sensu Cronquist, 1981). Further, many of the reports are isolated identifications (single species, a few compounds) and with few exceptions (Venkataraman, 1972), little attempt has been made to summarize these scattered data. Most recent chemotaxonomic discussions of the Hamamelidae have been placed within the broader context of angiosperm phylogeny in general (e.g., Gershenzon & Mabry, 1983; Gornall et al., 1979; Harborne, 1977; Harborne et al., 1975; Harborne & Mabry, 1982; Young, 1981; Young & Seigler, 1981).

To be sure, several families have been surveyed in detail, such as the Betulaceae (Wollenweber, 1975) and Ulmaceae (Giannasi, 1978), as have several genera, for example, *Fagus* (Giannasi & Niklas, 1981) with, in some cases, emphasis on different tissues such as wood chemistry, for example, Moraceae (Venkataraman, 1972). All have been helpful at their respective taxonomic levels but of limited use above the family level. Macromolecular data for the Hamamelidae, in the form of serological studies, are now available through the efforts of Fairbrothers and co-workers (Brunner & Fairbrothers, 1979; Petersen & Fairbrothers, 1979, 1983, 1985). However, many of the phytochemical correlations that do exist for the Hamamelidae still rest on earlier secondary metabolite surveys, primarily phenolics, and it is here that major emphasis continues (Egger & Reznik, 1961; Bate-Smith, 1962; Kubitzki & Reznik, 1966; Jay, 1968).

This discussion is intended to: (1) provide a summarized update of the earlier review by Mears (1973) in terms of some of the new classes of compounds discovered in the Hamamelidae in the past ten years and (2) to highlight several types of chemical data that have recently been

[1] My thanks to Patricia Holmgren (NY), Thomas Delendick (BKL), and S. B. Jones and Nancy Coile (GA) for providing extensive herbarium and cultivated samples of a number of the taxa studied in this work. My thanks also to Joscelyn Hill for technical assistance on the chemical studies. This work was supported by National Science Foundation grants DEB-8120515 and BSR-8120515.

[2] Department of Botany, University of Georgia, Athens, Georgia 30602.

ANN. MISSOURI BOT. GARD. 73: 417–437. 1986.

TABLE 1. Subclass Hamamelidae according to Cronquist (1981).

| Subclass Hamamelidae | Putative Relationships or Alternative Treatments[a] |
|---|---|
| Trochodendrales | Magnoliidae |
| Tetracentraceae | Hamamelidaceae; *Altingia, Liquidambar* |
| Trochodendraceae | |
| Hamamelidales | Trochodendrales |
| Cercidiphyllaceae | Hamamelidaceae (*Disanthus*) |
| Eupteleaceae | Cercidiphyllaceae, Platanaceae, Magnoliidae (Schisandraceae) |
| Platanaceae | |
| Hamamelidaceae | |
| Myrothamnaceae | |
| Daphniphyllales | |
| Daphniphyllaceae | Euphorbiaceae |
| Didymelales | — |
| Didymelaceae | Leitneriaceae, Euphorbiales, Thymelaceae |
| Eucommiales | |
| Eucommiaceae | Urticales, Hamamelidaceae, Magnoliales |
| Urticales | Malvales |
| Barbeyaceae | Malvales |
| Ulmaceae | Fagaceae, Betulaceae |
| Cannabaceae | Moraceae |
| Moraceae | |
| Cecropiaceae | Moraceae, Urticaceae |
| Urticaceae | Moraceae |
| Leitneriales | |
| Leitneriaceae | Didymelales |
| Juglandales | Rutales (Sapindales) |
| Juglandaceae | Anacardiaceae (Julianaceae) |
| Rhoipteleaceae | Myricaceae, Fagaceae, Betulaceae |
| Myricales | Juglandales, Fagales |
| Myricaceae | |
| Fagales | |
| Balanopaceae | |
| Fagaceae | |
| Betulaceae | *Trigonobalanus*, Fagaceae |
| Casuarinales | |
| Casuarinaceae | Betulaceae, Myricaceae |

[a] Taxa in this column represent alternative taxa in which the Hamamelidae (sensu Cronquist, 1981) have been placed by other authors (see text at right for references).

applied to systematic problems in the Hamamelidae, sensu Cronquist (1981). The latter Hamamelid concept is chosen primarily because of my personal familiarity with the system and its innate pedagogical convenience as a framework against which other treatments may be compared. It certainly has provided a focal point for the spirited systematic discussion over the definition of the Hamamelidae.

The historical development of the concept of the "Amentiferae" into the Hamamelidae has been described by Stern (1973). There are also several different treatments of the Hamamelidae (within larger angiosperm classifications) in contemporary systematics (Conquist, 1968, 1981; Thorne, 1983; Dahlgren, 1980; Takhtajan, 1954, 1969, 1980). Some of these undergo regular revisions (Dahlgren, 1977, 1980, 1983; Thorne, 1973, 1976, 1977, 1983). Others are more specific reviews of the Hamamelidae alone (Abbe, 1974; Endress, 1977; Meeuse, 1975) or of specific orders along with other putative relationships within the subclass (e.g., Berg, 1977). Merxmüller (1977) has succinctly commented on the relative merits of a number of these systems.

Depending on one's taxonomic predilection, the Hamamelidae contain the taxa shown in Table 1, the left-hand column of taxa representing Cronquist's treatment, the column to the right showing a selection of some other relationships suggested by other workers. Most authors concur that a basic "core" of taxa including the Hamamelidales and Fagales probably represent the true concept of Hamamelidae (and then perhaps conservatively only the type families). All the other orders (a number of which are monotypic or at least monogeneric) are moved with great frequency (and often with justifiable logic) to other subclasses, orders, or families and back again. The phytochemist is often at a loss as to which and how many taxa to sample to provide an adequate survey of what various taxonomists consider the Hamamelidae and related taxa.

## CHEMICAL REVIEW

If any group has exploited the use of phenolic compounds, surely it is the Hamamelidae. Since many taxa in Hamamelidae are woody, or essentially so, this is perhaps not unexpected and Cronquist (1977) attributed this characteristic phenolic synthesis, especially tannins, to a general chemical adaptation to herbivore deterrence by these compounds. Also, many unusual phenolic compounds are found in various members of this subclass (Mears, 1973) as in, for example, the Moraceae (Fig. 1; cf. Venkataraman, 1972). However, as Mears indicated, many of these compounds often are characteristic of only a few

species or a genus or a family and thus of little taxonomic help beyond that taxonomic level, especially in confirming or denying their position in the Hamamelidae. Alternatively, these compounds may be scattered throughout other disparate taxa in the angiosperms as a whole. The current known distribution of flavonoid biflavonyls is an example of such a class of compounds. In other cases, probably the majority, comprehensive surveys are lacking. Any heady phylogenetic relationship among plant taxa based on the presence or absence of a single or limited number of chemical characters often lasts only until the publication of the next phytochemical survey.

In general, tannins, proanthocyanidins (e.g., prodelphinidin), ellagic acid, and especially myricetin (and other vicinyl-hydroxylated compounds), are common to the Hamamelidae (Fig. 1). They have been used most commonly to separate the Hamamelidae (generally present) from the Magnoliidae (generally absent), although there are reports of their scattered occurrence in the Rosidae and Dilleniidae as well, with single isolated reports in the Asteridae and Liliopsida (although the Nymphaeales commonly have both ellagic acid and myricetin). A few species of the Magnoliidae also possess myricetin (Piperales, Laurales). The flavonoids in the Hamamelidae, while produced in large quantities, are often qualitatively simple flavonols (myricetin, quercetin, kaempferol). Some taxa possess glycoflavones, but with flavones generally low in number, at least by present survey data. O-methylation of flavonols and flavones in the Hamamelidae also seems to be low, with some exceptions (Betulaceae, Wollenweber, 1975; Moraceae, Venkataraman, 1972), but certainly not to the extent seen in other subclasses (e.g., Rosidae, Dilleniidae, Asteridae). Thus, the Hamamelidae possess a qualitatively conservative flavonoid complement, again at least by present surveys—perhaps the most significant caveat for such conclusions.

The occurrence of biflavonyls (Fig. 1) in *Casuarina* is unusual, as is their occurrence in a few other angiosperms. These flavonoid dimers are more consistently characteristic of gymnosperms and some lower tracheophytes (except ferns) and a moss or two. In angiosperms they occur in such disparate groups as Nandinaceae (Ranunculidae), Rhamnaceae, Euphorbiaceae, Thymelaceae, Ochnaceae, Clusiaceae (Guttiferae), Anacardiaceae, Burseraceae, Caprifoliaceae, and most recently have been found in the monocots, the Amaryllidaceae (Liliaceae). Their scattered distribution offers little systematic value at this time and may represent isolated parallel synthetic capabilities or perhaps only a lack of comprehensive surveys (Geiger & Quinn, 1975, 1982). More typical flavonoid monomers also occur commonly in *Casuarina* spp. consisting of a number of glycosidic variations based on a conservative aglycone complement of myricetin, quercetin, and kaempferol (Saleh & El-Lakany, 1979).

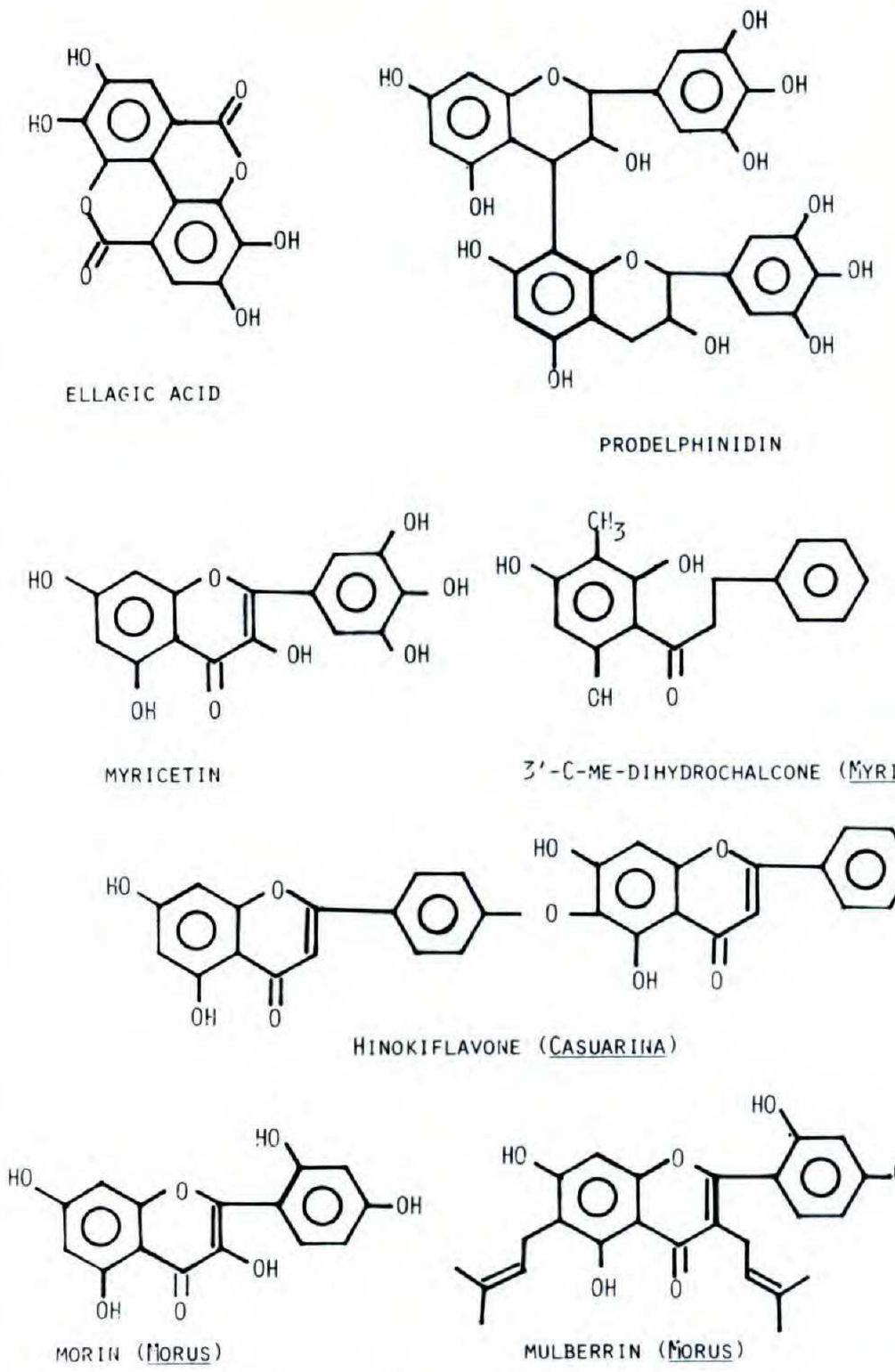
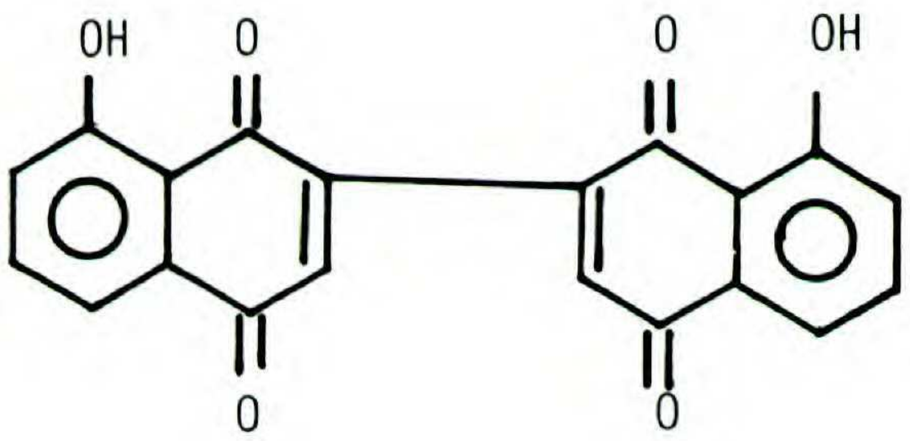



FIGURE 1. Examples of different types of phenolics in the Hamamelidae.

Quinones (Fig. 2) are found in several families in the Hamamelidae, especially the Juglandaceae, which produces the allelopathic agent, juglone, and several allied compounds such as bis-juglone (Gupta et al., 1972; Pardhasaradhi & Babu, 1978). Mixed phenolic-terpene (and sesquiterpene) quinones such as the aromatic naphthalenes of Ulmaceae heartwoods are known (Mears, 1973) along with some rare flavonol and flavanonol C-glycosides (Thomson, 1979; Hillis & Horne, 1966) and unusual C-methyl dihydrochalcones in *Myrica* (Malterud et al., 1977; Uyar et al., 1978). Fatty acid patterns in nut oils of *Carya* species have also been employed with systematic success (Stone et al., 1969).




3, 3'-BISJUGLONE

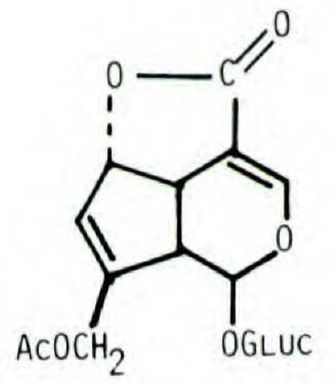
JUGLANS REGIA




LINOOL-9-O-GLUCOSIDE

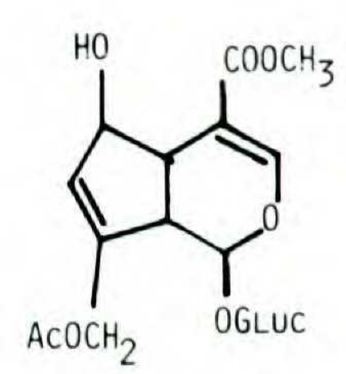
BETULA ALBA

**FIGURE 2. Quinone and monoterpene types in the Hamamelidae.**



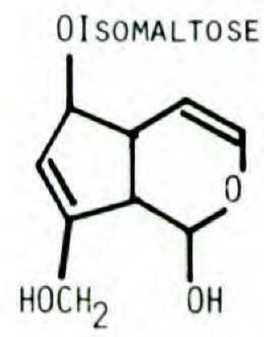

ASPERULOSIDE

DAPHNIPHYLLACEAE



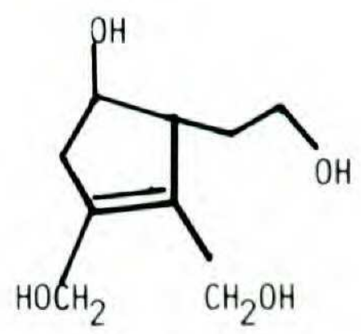

DAPHNIPHYLLOSIDE

DAPHNIPHYLLACEAE




ULMOSIDE

EUCOMMIACEAE




EUCOMMIOL

EUCOMMIACEAE

**FIGURE 3. Examples of iridoids found in some Hamamelidae.**

Several types of terpenes are found in the Hamamelidae. Typical monoterpenes are found in *Myrica* (Myricaceae) and these have been used for chemosystematic purposes (Halim & Collins, 1973). Unusual monoterpene glycosides (Fig. 2) occur in *Betula* (Tschesche et al., 1977). Monoterpene lactones or iridoids (Fig. 3) have also been found in the Hamamelidaceae (*Liquidambar*), Daphniphyllaceae (*Daphniphyllum*), Eucommiaceae (*Eucommia*), and Didymelaceae (*Didymeles*). Undoubtedly more will be discovered as specific surveys continue (El-Naggar & Beal, 1980; Kaplan & Gottlieb, 1982; Gershenzon & Mabry, 1983; Bianco et al., 1982). At this time the presence of iridoids in a few Hamamelidae further isolates this group from the Magnoliidae, which currently appear to lack them. Sesquiterpene lactones appear to be absent from the Hamamelidae as well but present in the Magnoliidae, providing yet another distinguishing character between the two subclasses.

In terms of nitrogen containing secondary products, glucosinolates appear to be absent from all Hamamelidae regardless of whose taxonomic scheme is used. Reports of cyanogenic glycosides (Fig. 7) rest mainly on color tests rather than extensive specific compound identifications. Such color tests have been reported for several families (Hamamelidaceae, Fagaceae, Juglandaceae). Complete identification is needed to clear this up especially in a biosynthetic sense (Hegnauer, 1973, 1977), but the few found are of the tyrosine-derived types.

Several types of alkaloids occur in the Hamamelidae (Fig. 4). Most are characteristic of a genus or two and of limited use due to their restricted occurrence, inadequate survey, or scattered occurrence in seemingly unrelated (at least not closely related) taxa (Mears, 1973). These include tropine types (pseudopelletierine) in *Ficus* (Moraceae) along with the tylophoric alkaloids, which also occur in *Tylophora* (Asclepiadaceae) and *Cryptocarya* (Lauraceae). A series of diterpene alkaloids (e.g., daphniphylline, daphnigraciline) have been reported from *Daphniphyllum gracile*, mostly from bark, and as yet are

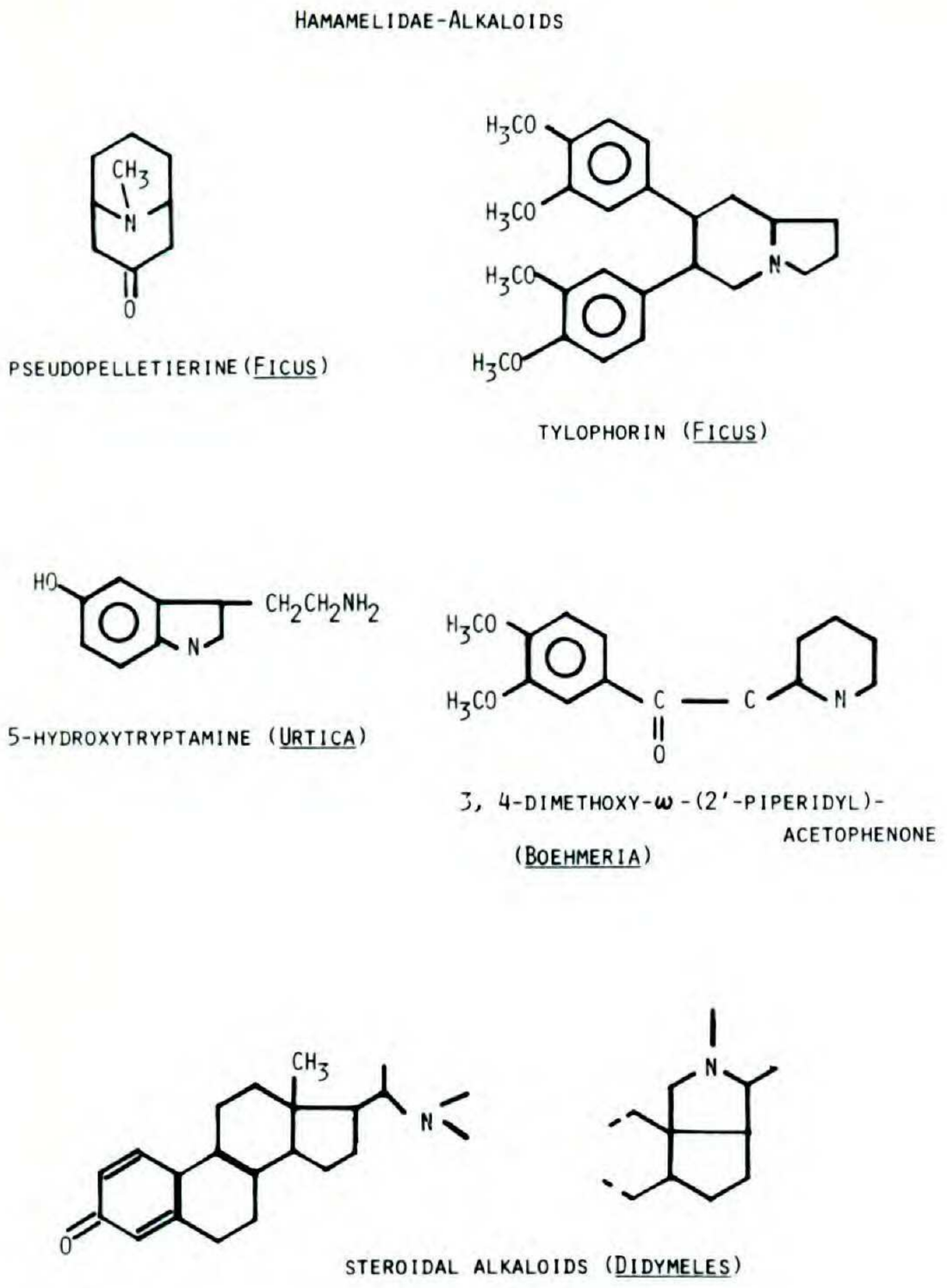



FIGURE 4. Alkaloids in the Hamamelidae.

taxonomically isolated in this occurrence (Grundon, 1977, 1981; Yamura et al., 1977, 1980). Steroid alkaloids have also been discovered in *Didymeles* (Ahond et al., 1980). The Hamamelidae generally appear to lack the tyrosine/phenylalanine-derived (benzyl-) isoquinoline alkaloids of the Magnoliidae-Caryophyllidae-Lilliidae (Liliopsida). In this way, the Hamamelidae are more like Rosidae-Dilleniidae-Asteridae, in which non-aromatic derived alkaloids (amino acids from TCA cycle or terpenoids) begin to predominate.

Obviously there is an unusual variety of secondary metabolites in the Hamamelidae, but comprehensive surveys are lacking. The few chemical studies of specific genera or families using single classes of compounds in detailed surveys have produced both interesting systematic results and grist for the chemist's mill, as indicated in the following discussion.

## CHEMOSYSTEMATIC STUDIES—MICROMOLECULAR

Some of the more recent comprehensive studies in the Hamamelidae have centered on the Ulmaceae. An early flavonoid study of *Ulmus* and several related genera (Bate-Smith & Richens, 1973) showed that flavonoid evolution in the genus probably proceeded by reduction in flavonoid types and content (mostly flavonols). Bate-Smith and Richens also noted that several other related genera differed in their possession of flavone compounds but did not pursue it further. Subsequent studies by Giannasi and Niklas (1977) suggested that a flavonoid dichotomy existed between *Ulmus* (flavonols) and *Celtis* (glycoflavones). A comprehensive flavonoid aglycone study (Giannasi, 1978) confirmed this dichotomy as shown in Table 2. This generic arrangement generally matches that of Grudzinskaya (1965), who had previously proposed two separate families (Ulmaceae and Celtidaceae) rather than the more common treatment of two subfamilies (cf. Giannasi, 1978, for discussion and references). Later, SEM pollen analyses by Zavada supported Grudzinskaya's treatment (Zavada, 1983). As Zavada indicated, these data, along with fossil evidence (Zavada & Crepet, 1981), suggest that the two subfamilies have had a separate phylogenetic history since Eocene times and perhaps earlier.

TABLE 2. Generic distribution of flavonoids in the Ulmaceae (Giannasi, 1978).

| | Flavonoids | Affinity[a] |
|---|---|---|
| **Flavonols** | | |
| 1. | *Ampelocera* | C |
| 2. | *Aphananthe* | C |
| 3. | *Barbeya* | (—) |
| 4. | *Chaetoptelea* | (—) |
| 5. | *Hemiptelea* | U |
| 6. | *Holoptelea*[b] | U |
| 7. | *Mirandaceltis* | (—) |
| 8. | *Phyllostylon* | U |
| 9. | *Planera* | U |
| 10. | *Ulmus* | U |
| 11. | *Zelkova* | U |
| 12. | *Gironniera*: *Galumpita*[c] | |
| **Glycoflavones** | | |
| 12a. | *Gironniera*: *Gironniera* | C |
| 13. | *Celtis* | C |
| 14. | *Chaetachme* | C |
| 15. | *Lozanella* | C |
| 16. | *Parasponia* | C |
| 17. | *Plagioceltis* | (—) |
| 18. | *Pteroceltis* | C |
| 19. | *Trema* | C |

[a] According to Grudzinskaya (1965); C = Celtoid, U = Ulmoid, (—) = not considered by Grudzinskaya.
[b] Data from Bate-Smith and Richens (1973).
[c] Placed in *Aphananthe* by some authors.

TABLE 3. Leaf flavonoid[a] distribution in genera of the Juglandaceae (Giannasi & Niklas, unpubl. data).

| | Flavonols | | | Fla-vones | Glyco-flavones | Flavanonols | | Phenolics | |
|---|---|---|---|---|---|---|---|---|---|
| Taxon[b] | M | Q | K | A | A | DQ | DK | EA | GA |
| *Platycarya* (1) | | + | + | + | | + | | + | |
| *Pterocarya* (6) | | + | + | + | | + | | + | |
| *Alfaroa* (3) | | + | + | + | | + | | + | |
| *Juglans* A. (7) | + | + | + | + | | + | + | + | + |
| B. (6) | | + | + | + | | + | | + | + |
| *Carya* A. (6) | + | + | + | + | | + | | + | |
| B. (6) | | + | + | + | ? | + | | + | |
| *Oreomunnea* A. (1) | + | + | + | + | | + | | + | |
| B. (1) | | + | + | + | | + | | | |
| *Engelhardia* (3) | + | + | + | + | | + | | + | |

[a] Abbreviations: M = myricetin, Q = quercetin, K = kaempferol, A = apigenin, DQ = dihydroquercetin, DK = dihydrokaempferol, EA = ellagic acid, GA = gallic acid derivatives, + = present, ? = not completely confirmed.

[b] Number in parentheses indicates number of species examined in each genus.

The Juglandaceae are another recently studied group. Cronquist (1981) placed them in their own order along with the Rhoipteleaceae. Thorne (1983), however, placed the family in his superorder, Rutiflorae, suborder Juglandineae, not far from the Anacardiaceae (suborder Rutinae); Dahlgren (1980) places them in his Rosiflorae along with most of the Hamamelidae (sensu Cronquist, 1981).

In a recent flavonoid study of the Juglandaceae (Giannasi & Niklas, unpubl. data) it was found that mostly common flavonol glycosides, including those of myricetin, quercetin, and kaempferol along with two flavanols were produced in the leaves of the Juglandaceae (Table 3). Flavones and glycoflavones apparently are absent, or if present occur in trace amounts that are difficult to recover. The several genera examined may be separated into two major groups based on the presence or absence of myricetin glycosides, as shown in Table 3. All of the genera that produce myricetin occur in the New World. If the presence of myricetin is considered a primitive character, then one-character chemotaxonomy would suggest that the family may have originated in temperate North America. Indeed, the only exception to this is the Asian genus *Engelhardia,* which does produce myricetin. However, this apparently "Asian" taxon producing myricetin was widely represented in North America during Eocene times (Dilcher et al., 1976; Crepet et al., 1975), its current remaining Asian "endemism" being a secondarily derived or simply fortuitous relictual distribution. The myricetin marker compounds also proved to be useful in confirming the taxonomic affinities of a Miocene fossil leaf compression as that of a *Juglans* and its putative relationship to North American taxa.

The presence of this rather conservative leaf flavonoid complement in the Juglandaceae certainly allows the family to lie comfortably within the Hamamelidae. A suggested relationship of the Juglandaceae with or close to the Anacardiaceae, based primarily on the presence of ellagic acid and myricetin in both taxa, does not seem strong at this point, especially, when compared with the large number of unusual flavonoid types found in the Anacardiaceae (including Julianiaceae) by Young (1976, 1979). The Anacardiaceae, for example, possess anthochlor pigments, methylated flavonols, and 5- and 7-deoxyflavonoids not found in the Juglandaceae. Also, the lack of 5-methoxy flavonoids in the Anacardiaceae suggests that the Juglandaceae (which do possess them) are more compatibly retained in the Hamamelidae (at this time). The presence of biflavonyls in the Anacardiaceae absolutely sets this family apart from the Juglandaceae in which they are unknown.

A comprehensive survey of leaf bud flavonoid exudates has been carried out on the Betulaceae, including the genera *Betula, Alnus,* and *Ostrya* by Wollenweber (1975). All three genera could be distinguished on the basis of their flavonoids, and considerable interspecific flavonoid differences were observed within each genus. What was most interesting was the very large number of O-methylated flavonols that occurred in these genera, as well as a few methylated flavones, a flavonoid character of advancement (including

6-hydroxylation) observed only in some Rosidae and Asteridae, but not in other Fagales based on published surveys. Strangely enough, myricetin glycosides are not reported in the bud scales but commonly occur in the mature leaves along with several of the flavone and flavonol types cited for bud scales (Giannasi, unpubl. data). The family, therefore, seems to possess some advanced (or specialized) biosynthetic capability (O-methylation, 6-substitution) within the Hamamelidae, although more primitive myricetin glycosides do appear in the leaves of some species of *Betula*.

Beyond these few comprehensive surveys, most discussions are based on older broad surveys of aglycone hydrolysis in angiosperms (Bate-Smith, 1962; Lebreton, 1965; Jay, 1968) in which the flavonol aglycones are reported. A few (Kubitzki & Reznik, 1966; Gurni & Kubitzki, 1981; Egger & Reznik, 1961) do identify other flavonoids and their glycosides. Flavones or other compounds generally are not cited even though they are present (see below), which can give rise to spurious interpretations as we shall see.

## Chemosystematic Studies—Macromolecular

Available macromolecular data on taxa in the Hamamelidae emanate from the efforts of Fairbrothers and colleagues (Brunner & Fairbrothers, 1979; Petersen & Fairbrothers, 1979, 1983) and have been obtained at several taxonomic levels.

In a serological study of the Corylaceae (Brunner & Fairbrothers, 1979), serological affinities of representative taxa from *Alnus, Betula, Carpinus, Corylus,* and *Ostrya* were examined. Using four serological techniques the genera could be divided into three major groups: (1) *Alnus,* (2) *Betula,* and (3) *Carpinus, Corylus,* and *Ostrya. Betula* proved to be the most serologically isolated taxon of the five but showed closest affinities with *Alnus. Alnus,* though distinct, was most similar to *Corylus* of Group 3. Overall similarities between all five genera suggest that they be retained within a single family (as tribes corresponding to the serological groupings) rather than elevating Group 3 to familial status, that is, Corylaceae.

In a second study (Peterson & Fairbrothers, 1979), an attempt was made to determine if the Juglandaceae, Myricaceae, and Fagaceae were closest to a Hamamelid origin (Cronquist, 1981; Takhtajan, 1969; Hutchinson, 1959) or if the Juglandaceae and Myricaceae are of a Rutalean origin near the Anacardiaceae (Thorne, 1976) with only the Fagaceae and Hamamelidaceae retained within the Hamamelidae, or one of several other possibilities mentioned by the other workers. Serology indicates that the Fagaceae and Myricaceae are closely related and show close similarity with the Juglandaceae as suggested by Cronquist (1981) and Takhtajan (1980). Little similarity between these three families and the Anacardiaceae was observed, thus failing to support such a relationship. As mentioned earlier, our own flavonoid surveys of the Juglandaceae also fail to support any strong relationship with the Anacardiaceae.

In a third study, an attempt was made to deal with one of the peripheral taxa in the Hamamelidae (sensu Cronquist, 1981), the Leitneriales, a monotypic order (Peterson & Fairbrothers, 1983) placed close to the Hamamelidales and near the Fagales-Myricales-Juglandales by Cronquist. In fact, serology suggests that the strongest affinity of the Leitneriales lies with *Ailanthus* and *Picrasma* of the Simaroubaceae and thus it is of Rutalean origin rather than of Hamamelid origin.

With the limited macromolecular data available, support is given in various examples for a broad concept of the Betulaceae, a solid relationship of Hamamelidales-Fagales-Myricales within, or as, the Hamamelidae (or at least as a natural taxonomic unit regardless of whose treatment is followed) and the removal of a peripheral group, Leitneriales, to the vicinity of the Simaroubaceae (Petersen & Fairbrothers, 1985). The results are encouraging and we can only hope that further studies will be conducted.

## Hamamelidae—Current Surveys

I also undertook a limited survey of phenolics and flavonoids in the Hamamelidae both to check the results of earlier studies, especially the monumental work by Bate-Smith (1962) as well as others (Lebreton, 1965; Jay, 1968) and to add data for a few additional taxa where possible. The methodology employed was that of Giannasi (1978).

It was observed in the earlier studies that usually only the presence of flavonol aglycones was reported although from my own studies it was often obvious that other flavonoid classes were also present. My own studies further indicated that these other compounds were glycoflavones and flavones, in addition to the flavonols. Thus, the earlier studies, which emphasized only fla-

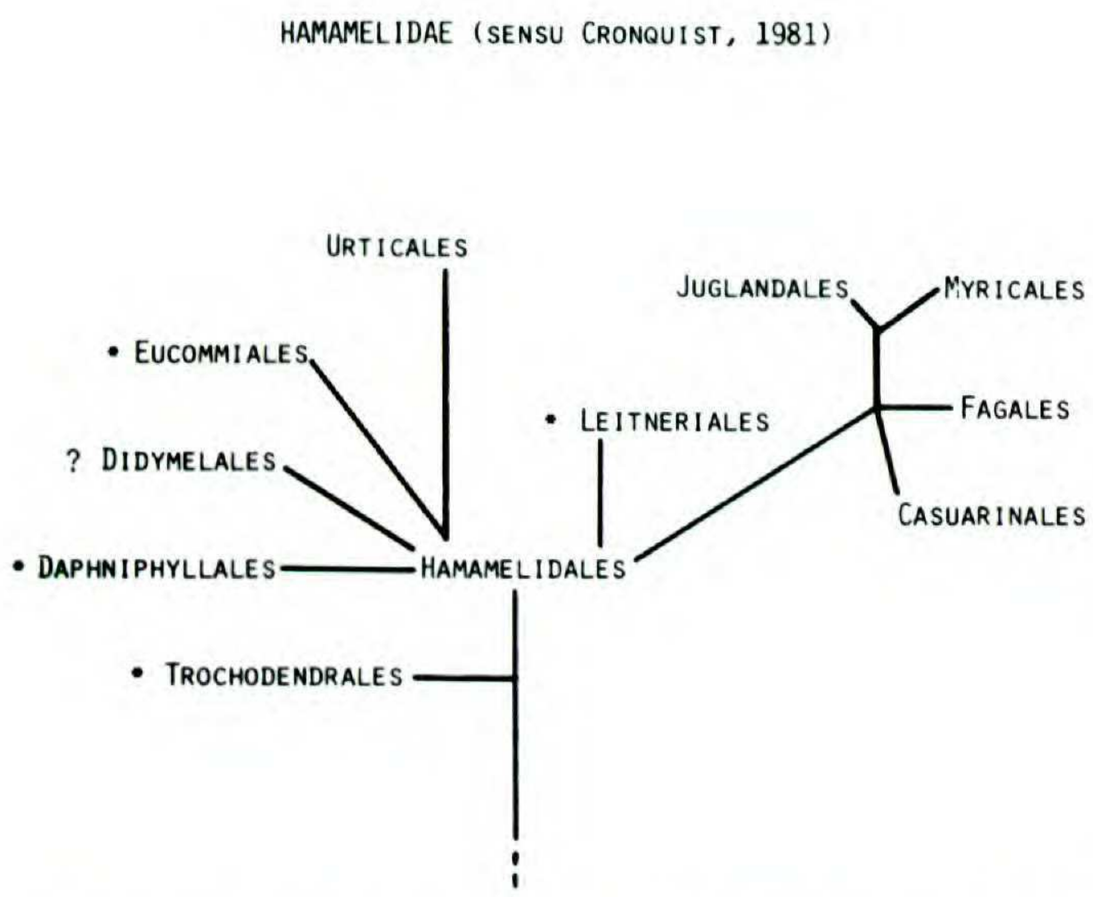



FIGURE 5. Putative phylogeny of the Hamamelidae (sensu Cronquist, 1981). Asterisk indicates lack of ellagic acid and myricetin compounds.

TABLE 4. Glycoflavone distribution in some Hamamelidae.

| Family | Genera |
|---|---|
| **Glycoflavones** | |
| Cecropiaceae | *Cecropia, Pourouma* |
| Hamamelidaceae | *Sinowilsonia* |
| Moraceae | *Helicostylis, Ficus* |
| Urticaceae | *Urtica*(?) |
| **Glycoflavones/Flavonols** | |
| Hamamelidaceae | *Hamamelis* |
| Leitneriaceae | *Leitneria*(?) |
| Moraceae | *Helicostylus, Cudrania* |
| **Glycoflavones/Simple Flavones** | |
| Moraceae | *Ficus, Dorstenia, Broussonettia* |
| **Glycoflavones/Flavones/Flavonols** | |
| Cannabaceae | *Humulus* |

vonols, inadvertently left out other flavonoids of considerable potential taxonomic importance, especially when comparing different subclasses, as we shall see below. In my own survey I was unable to confirm the presence of some aglycones cited in earlier studies of the same taxa. In some of these taxa I found additional aglycones not previously noted. In these cases such differences are most likely attributable to natural infraspecific variation exhibited by some taxa. The presence of flavonol aglycones (myricetin, quercetin, kaempferol) often depended on the number of collections sampled, sometimes requiring a composite flavonoid score from several or more collections to characterize a taxon. For example, in the extensive literature survey by Gornall et al. (1979) the Casuarinaceae were said to lack myricetin (Bate-Smith, 1962), but a contemporary detailed survey (Saleh & El-Lakany, 1979) as well as my own survey clearly document the presence of myricetin and the relative small quantities of biflavonyls produced in the leaves of *Casuarina* species. Therefore, all of these general surveys, including my own, that included taxa not sampled previously, must be considered provisional. Nevertheless, some correlations may suggest several phyletic trends in the evolution of the Hamamelidae and among its related subclasses.

If we consider Cronquist's phylogenetic treatment of the Hamamelidae (Fig. 5) we find that the "core" orders of the subclass possess myricetin and ellagic acid. A summarization of such data (Table 6) at the ordinal level suggests a "backbone" group consisting of the Hamamelidales-Fagales-Juglandales-Myricales-Casuarinales-Urticales. Similarly, many of the "peripheral" orders such as the Trochodendrales, Daphniphyllales, Eucommiales, and Leitneriales, whose presence in the Hamamelidae has been debated, lack both ellagic acid and myricetin, a fact confirmed by earlier and present studies. Care must be taken in using these generalizations, however, since not all species within a genus (e.g., *Myrica*) possess these characters although most do (Table 5), nor do all families within the "core" orders characterized by these constituents possess them (Table 6; e.g., Urticales, Hamamelidales). Indeed, in dealing with genera that may contain several hundred taxa, existing studies are certainly provisional.

Despite these caveats, several correlations and resultant hypotheses for the taxonomic grist mill are warranted based on current evidence. For example, the presence of myricetin and ellagic acid is considered a primitive chemical character (Bate-Smith, 1962; Harborne 1977). This also suggests that the peripheral orders of the Hamamelidae that *lack* these chemical characters may represent (1) a separate subclass, but closely related to the "core" Hamamelidae, and/or (2) a group of taxa exhibiting a combination of ancient or derived but parallel morphotypes, and thus, (3) that, one or more of these orders, while showing the general Hamamelid syndrome of anemophily, etc., perhaps may belong in other subclasses (see Table 1). Tiffney (1986) also indicated that there is little or no overall phyletic correlation in fruit dispersal mechanisms in the

TABLE 5. Compound distribution in taxa of the Hamamelidae sampled for phenolics and flavonoids.[a]

| Taxon | Phenolics | | Flavonols | | | Flavones | | | | Glycoflavones | | | Flava-nonols | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | GA | EA | M | Q | K | A | L | T | D | A | L | C | DQ | DK |
| Trochodendraceae | | | | | | | | | | | | | | |
| *Trochodendron araliensis* | | | | + | + | | | | | | | | | |
| Tetracentraceae | | | | | | | | | | | | | | |
| *Tetracentron sinense* | | | | + | + | | | | | | | | + | |
| Cercidiphyllaceae | | | | | | | | | | | | | | |
| *Cercidiphyllum japonicum* | + | | + | | + | + | | | | | | | | |
| Eupteleaceae | | | | | | | | | | | | | | |
| *Euptelea pleiospermum* | | | | | + | + | | | | | | | | |
| *E. polyandra* | | | | | + | + | | | | | | | | |
| Platanaceae | | | | | | | | | | | | | | |
| *Platanus acerifolia* | | | | | + | + | | | | | | | | |
| *P. occidentalis* | | | | | + | + | + | | | | | | | |
| Hamamelidaceae | | | | | | | | | | | | | | |
| *Corylopsis spicata* | | | + | + | + | + | | | | | | | | |
| *C. pauciflora* | | | + | + | + | + | | | | | | | | |
| *C. sinensis* (2 vars.) | + | | + | | + | + | | | | | | | | |
| *Distylium lepidotum* | + | | + | | + | + | | | | | | | + | + |
| *D. racemosum* | | | | | + | + | | | | | | | | |
| *Fortunearia sinensis* | | | | | + | + | | | | | | | | |
| *Fothergilla major* | + | | + | + | + | + | | | | | | | + | |
| *F. gardenii* | + | | + | ? | + | + | | | | | | | + | |
| *Hamamelis vernalis* | + | | + | + | + | + | | | | | | | | |
| *H. virginiana* | (+) | | + | + | + | + | | | | + | + | | ? | |
| *H. macrophylla* | + | | | ? | | + | | | | + | + | | ? | |
| *Liquidambar styraciflua* | | | + | + | + | + | | | | | | | (+) | |
| *L. formosa* | | | | + | + | + | | | | | | | (+) | |
| *Loropetalum chinense* | | | + | + | + | + | | | | | | | (+) | |
| *Sycopis sinensis* | (+) | | + | + | + | + | | | | | | | | |
| *Parrotia persica* | | | | | + | + | | | | | | | | |
| *Sinowilsonia henryii* | + | | + | | | | | | | + | + | | | |
| Myrothamnaceae | | | | | | | | | | | | | | |
| *Myrothamnus flabellifolium* | + | | + | + | + | + | | | | ? | ? | | | |
| Daphniphyllaceae | | | | | | | | | | | | | | |
| *Daphniphyllum teigmensis* | | | | ? | ? | + | + | | | | | | | |
| *D. glaucescens* | | | | ? | ? | + | + | | | | | | | |
| *D. calycinum* | | | | ? | ? | + | + | | | | | | | |
| Didymelaceae | | | | | | | | | | | | | | |
| *Didymeles* spp. | (unknown?) | | | | | | | | | | | | | |
| Eucommiaceae | | | | | | | | | | | | | | |
| *Eucommia ulmoides* | | | | + | + | | | | | | | | | |
| Barbeyaceae[b] | | + | | + | + | | | | | | | | | |
| Ulmaceae[b] | | + | + | + | + | + | + | | | + | + | | | |
| Cannabaceae[c] | | | | | | | | | | | | | | |
| *Humulus americana* | | | + | + | + | | | | | | | | | |
| *H. japonicus* | | | | + | | + | + | | | + | + | | | |

TABLE 5. Continued.

| Taxon | Phenolics | | Flavonols | | | Flavones | | | | Glycoflavones | | | Flava-nonols | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | GA | EA | M | Q | K | A | L | T | D | A | L | C | DQ | DK |
| **Moraceae** | | | | | | | | | | | | | | |
| *Broussonettia papyrifera* | | | | | | + | + | | | + | + | ? | | |
| *Cudrania tricuspidata* | | | | + | + | | | | | + | | | + | |
| *Dorstenia foetida* | | | | | | | + | | | + | + | | | |
| *Fatoua specium* | | | | + | | | | | | | | | | |
| *Ficus aurea* | | | | + | + | | | | | | | | | |
| *F. benjamina* | | | | | + | | | | | | | | | |
| *F. brevifolia* | | | | + | + | | | | | | | | | |
| *F. caprifolia* | | | | | | | | | | + | | | | |
| *F. caprica* | | | | | | | | | | + | | | | |
| *F. citrifolia* | | | | + | + | | | | | + | | | | |
| *F. gemina* | | | | | | + | + | + | | + | + | | | |
| *F. laevigata* | | | | + | + | | | | | | | | | |
| *F. llewellynii* | | | | + | + | | | | | | | | | |
| *F. macrophylla* | | | | + | + | | | | | | | | | |
| *F. nitidifolia* | | | | + | + | | | | | | | | | |
| *F. pumila* | | | | | + | | | | | | | | | |
| *F. webbiana* | | | | | + | | | | | | | | | |
| *Helicostylis elegans* | | | | + | | | | | | + | + | | | |
| *H. scabra* | | | | | | | | | | + | + | | | |
| *Morus alba* | | | | + | + | | + | | | | | | | |
| *M. rubra* | | | | + | + | | | | | | | | | + |
| **Cecropiaceae** | | | | | | | | | | | | | | |
| *Cecropia peltata* | | | | | | | | | | + | + | | | |
| *Pourouma phaeotricha* | | | | + | + | | | | | | | | | |
| *P. palmata* | | | | | | | | | | + | + | | | |
| **Urticaceae** | | | | | | | | | | | | | | |
| *Boehmeria cylindrica* | | | | + | + | | | | | | | | | |
| *Laportea canadensis* | | | + | + | + | + | | | | | | | | |
| *Urtica dioica* (2 vars.) | | | | + | + | | | | | ? | | | | |
| **Leitneriaceae** | | | | | | | | | | | | | | |
| *Leitneria floridana* | | | + | + | + | + | + | | | ? | | | | |
| **Juglandaceae**[d] | + | + | + | + | + | | | | | ? | | | | + |
| **Rhoipteleaceae** | | | | | | | | | | | | | | |
| *Rhoiptelea chiliantha* | | + | + | + | + | | | | | | | | | |
| **Myricaceae** | | | | | | | | | | | | | | |
| *Myrica asplenifolia* (= *Comptonia perigrina*) | | + | + | + | + | | | | | | | | | |
| *M. cerifera* | | | + | + | + | | | | | | | | | ? |
| *M. gale* | | + | + | + | + | | | | | | | | | |
| *M. heterophylla* | | | + | + | + | | | | | | | | | |
| *M. inodora* | | | + | + | + | | | | | | | | | + |
| *M. rubra* | | | + | + | + | | | | | | | | | + |
| *M. serrata* | | + | + | + | + | | | | | | | | | + |
| **Balanopaceae** | | | | | | | | | | | | | | |
| *Balanops* | (unknown ?) | | | | | | | | | | | | | |

TABLE 5. Continued.

| Taxon | Phenolics | | Flavonols | | | Flavones | | | | Glycoflavones | | | Flava-nonols | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | GA | EA | M | Q | K | A | L | T | D | A | L | C | DQ | DK |
| Fagaceae | | | | | | | | | | | | | | |
| *Quercus*[e] | | + | | + | + | + | + | | | + | | | + | |
| *Castanea*[e] | | + | | + | | | | | | | | | + | |
| *Fagus*[f] | | | | + | + | | | | | | | | | |
| Betulaceae[g] | | | | | | | | | | | | | | |
| *Betula* | + | | + | + | + | + | + | | | (+) | | | | |
| Casuarinaceae[h] | | | | | | | | | | | | | | |
| *Casuarina equisetifolia* | + | | + | + | + | | | | | | | | | |
| *C. glauca* | + | | + | + | + | | | | | | | | + | |
| *C. cunninghamia* | + | | + | + | + | | | | | | | | + | |

[a] Abbreviations: GA = gallic acids, EA = ellagic acids, M = myricetin, Q = quercetin, K = kaempferol, A = apigenin, L = luteolin, T = tricin, D = diosmetin, C = chrysoeniol, DQ = dihydroquercetin, DK = dihydro-kaempferol, + = present, (+) = trace amounts or occasionally present, ? = not completely confirmed.

[b] See Giannasi (1978) for detailed distributions and Table 2. Also see Bate-Smith and Richens (1973).

[c] See also Clark and Bohm (1979).

[d] See Table 3 (Giannasi & Niklas, unpubl. data).

[e] See Niklas and Giannasi (1978).

[f] See Giannasi and Niklas (1981).

[g] See also Wollenweber (1975). Present study also includes some methylated and/or 6-substituted flavones and flavonols as per Wollenweber.

[h] See also Saleh and El-Lakany (1979).

Hamamelidae and similar suggestions may be gleaned from discussion of pollination mechanisms (Whitehead, 1969). Indeed, protein serology (Petersen & Fairbrothers, 1983, 1985) suggests that the affinities of the Leitneriales, for example, lie near or within the Simaroubaceae (Rosidae, sensu Cronquist, 1981). Any one of the alternatives is possible for each of these peripheral orders, especially since most of these orders are monotypic or at least monogeneric. Often there are fewer intermediates that might more clearly suggest more direct interordinal relationships.

In addressing the flavonol bias of some earlier literature, it may be observed from Table 5 that the glycoflavones represent a second major class of flavonoids occurring in the Hamamelidae. As indicated in Tables 4 and 5, these flavonoids, either exclusively or in combination with flavonols and/or flavone O-glycosides, characterize a number of species and genera in various families. This is especially striking in the Daphniphyllaceae in which glycoflavones occur exclusively; a character state considered advanced or derived over the presence of flavonols alone or the intermediate state of flavonols/glycoflavones (e.g., Leitneriaceae). The same trends (i.e., flavonols → glycoflavones) may also be observed at various taxonomic levels within orders (Urticales), families (Moraceae), and genera (*Hamamelis, Ficus*) and thus may represent the major chemical trend of advancement within the subclass. This contrasts with earlier literature, which state that flavonols characterize the Hamamelidae as a whole, implying more of a conservative flavonoid capability than really exists. Exceptions do exist, as in the Betulaceae, for example. In *Alnus, Ostrya,* and especially *Betula,* flavonoids from bud scale excretions contain a large number of variously methylated and 6-substituted derivatives of the flavonols quercetin and kaempferol and to a much lesser degree flavones (apigenin) and flavanones (naringenin). Many of these compounds also occur in the leaves of *Betula* species (Giannasi, unpubl. data) along with the more archaic myricetin glycosides, which apparently do not occur in the bud excretions. Thus, the Betulaceae have retained primitive characters (flavonols) along with specialized characters (6-substitution, 6-methylation, flavones). These highly specialized flavonoids apparently do not commonly occur in the related members of the Fagaceae that have been surveyed (Niklas & Giannasi, 1978; Giannasi & Niklas, 1981; Gian-

TABLE 6. Summary distribution of phenolics and flavonoids in the Hamamelidae (taxonomy follows that of Cronquist, 1981).[a]

| Taxon | Phenolic and Flavonoid Distributions[b] | | | | | | | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | EA | GA | PC | CH | M | Q | K | R | IR | DHF | F6L | GF | A | L | T | AM1 | AM2 | LM1 | LM2 | PF | BF | FV | F5M |
| Trochodendrales | | | | | | | | | | | | | | | | | | | | | | | |
| Tetracentraceae | | | | + | | + | (+) | | (+) | (+) | | | | | | | | | | | | | |
| Trochodendraceae | | | + | | | + | + | | | | | | | | | | | | | | | | |
| Hamamelidales | | | | | | | | | | | | | | | | | | | | | | | |
| Cercidiphyllaceae | + | + | | + | | + | + | | | | | | | | | | | | | | | | |
| Eupteleaceae | | | | + | + | + | + | | | | | | | | | | | | | | | | |
| Platanaceae | | | + | | + | + | + | | | | | | + | | | | | | | | | | |
| Hamamelidaceae | + | + | + | | + | + | + | | | + | | + | + | | | | | | | | | | |
| Myrothamnaceae | + | | | + | | + | + | | | | | | | | | | | | | | | | |
| Daphniphyllales | | | | | | | | | | | | | | | | | | | | | | | |
| Daphniphyllaceae | | | | | | ? | ? | | | | | | + | + | | | | | | | | | |
| Didymelales | | | | | | | | | | | | | | | | | | | | | | | |
| Didymelaceae | (unknown ?) | | | | | | | | | | | | | | | | | | | | | | |
| Eucommiales | | | | | | | | | | | | | | | | | | | | | | | |
| Eucommiaceae | | | | + | | + | + | | | | | | | | | | | | | | | | |
| Urticales | | | | | | | | | | | | | | | | | | | | | | | |
| Barbeyaceae | | | | | | + | + | | | | | | | | | | | | | | | | |
| Ulmaceae | + | | + | | + | + | + | | | + | | + | | | | | | | | | | | |
| Cannabaceae | | | (+) | + | (+) | + | + | | | + | | + | + | | | | | | | + | | + | |
| Moraceae | | | + | + | + | + | + | | (+) | + | | + | + | + | (+) | | | | | + | | + | |
| Cercropiaceae | | | (+) | | | | | | | | | + | | | | | | | | | | | |
| Urticaceae | | | + | | + | + | + | | | | | + | | + | | | | | | | | | |
| Leitneriales | | | | | | | | | | | | | | | | | | | | | | | |
| Leitneriaceae | | | | | + | + | + | | | | | | + | + | | | | | | | | | |
| Juglandales | | | | | | | | | | | | | | | | | | | | | | | |
| Juglandaceae | + | | + | | + | + | + | | | + | | ? | | | | | | | | | | + | + |
| Rhoipteleaceae | + | | | | + | + | + | | | | | | | | | | | | | | | | |
| Myricales | | | | | | | | | | | | | | | | | | | | | | | |
| Myricaceae | + | | + | + | + | + | (+) | | | | | | | | | | | | | | | | |

TABLE 6. Continued.

| Taxon | Phenolic and Flavonoid Distributions[b] | | | | | | | | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | EA | GA | PC | CH | M | Q | K | R | IR | DHF | GF | F6L | LM2 | PF | BF | FV | AM1 | T | L | A | AM2 | LM1 | F5M | |
| Fagales | | | | | | | | | | | | | | | | | | | | | | | | |
| Balanopaceae | (unknown ?) | | | | | | | | | | | | | | | | | | | | | | | |
| Fagaceae | + | | + | + | + | + | + | | + | + | | + | + | | | | + | | + | + | + | | | |
| Betulaceae | ? | | + | + | + | + | + | | | + | | + | | | | | + | | + | + | + | | | |
| Casuarinales | | | | | | | | | | | | | | | | | | | | | | | | |
| Casuarinaceae | + | | (+) | | + | + | + | | | | | | | | + | | | | | | | | | |

[a] See text and Table 5 for chemical authorities, especially Bate-Smith (1962), Kubitzki and Reznik (1966), and Gornall et al. (1979).
[b] Compound identification: EA = ellagic acid, GA = gallic acid, PC = proanthocyanidins, CH = chalcones/dihydrochalcones, M = myricetin, Q = quercetin, K = kaempferol, R = rhamnetin, IR = isorhamnetin, DHF = dihydroflavonols, F6L = flavonol-6 or 8 substituted, GF = glycoflavones, A = apigenin, L = luteolin, T = tricin, AM1 = 7.4′ OMe-apigenin-1, AM2 = 6/8 OMe-apigenin-2, LM1 = 3′4′7 OMe-luteolin-1, LM2 = 6-OH/OMe-luteolin-2, PF = prenylflavonoids, BF = biflavonyls, FV = flavanones, F5M = 5-OMe flavonoids.

nasi, unpubl. data). Therefore, at this time, these compounds in the Betulaceae seem to represent a unique event in the Hamamelidae.

Also notable is the occurrence of a large number of flavones (and a few xanthones) that are substituted at various positions by isoprene (5C) units rather than sugars, methoxy or sulfate units in the root bark of Moraceae (e.g., Nomura et al., 1976, 1977, 1978a, 1978b; Konno et al., 1977; Deshpande et al., 1973). Similarly substituted flavones (or flavanone or flavanonol analogues) also are found in the Rosidae (Fabaceae, Rutaceae) and Asteridae (Asteraceae) and thus are not unique to the Hamamelidae but are unique within a single family of the Hamamelidae. That most of these prenylated flavones in the Hamamelidae have thus far been isolated only from root bark tissue of the Moraceae further emphasizes the possibility of inter-tissue chemical differences, which eventually must be considered in chemical studies (Gornall et al., 1979). Indeed, leaf flavonoids, in this case glycoflavones, seem to be "normally" substituted with C-glycosyl sugars. The occurrence of these prenylated flavones in the Moraceae may represent simply an isolated specialization in the Hamamelidae, as is suggested by the isolated occurrence of biflavonyls in the Casuarinaceae. The latter compounds, too, are unique to the Casuarinaceae in the Hamamelidae but do occur in other subclasses (Rosidae, Dilleniidae). The possibility that the Moraceae do not belong in the Hamamelidae is also possible (see p. 433 for additional discussion). Their scattered occurrence within the angiosperms makes them of questionable taxonomic value at this time.

## HAMAMELIDAE—GENERAL CONSIDERATIONS

The Hamamelidae, in terms of their phenolics, seem to represent a primitive group in the general presence of proanthocyanidins, ellagic acid, myricetin compounds, and a general conservatism in other flavonoids. Variation is based on glycosylation patterns of a few simple flavonol and flavone types with a moderate substitution, in some cases, by the evolutionarily intermediate glycoflavones. Proanthocyanidins, ellagic acid, and myricetin flavonoids are considered primitive chemical characters and are often found to be characteristic of woody plants (Bate-Smith, 1962; Harborne, 1977). Thus, the Hamamelidae are distinct from the Magnoliidae and Liliopsida, which generally lack one or more of these compounds. Yet these same Hamamelid characters

are found to some degree among the Rosidae and Dilleniidae, suggesting a more than casual relationship. The predominance of these chemical characters in the Hamamelidae, however, act more as a mark of exclusion of other subclasses because of the preponderance of their occurrence in the Hamamelidae, rather than as any absolute qualitative distinction (or requirement of "primitiveness"). Since non-chemical characters are often used similarly, such correlations among chemical characters are probably just as legitimate. For example, recent ultrastructural studies of phloem sieve-tube plastids show the uniform presence of the S-type in the Hamamelidae (with the exception of *Ulmus* species), but the same type is also found in some Magnoliidae and Rosidae (Behnke, 1973, 1977).

The problem of production of rather rare and unusual compounds in the Hamamelidae (especially the Moraceae), which are of restricted taxonomic use, has already been mentioned. Difficulties arise in the use of chemical data only in their simple distributional form without any consideration of the classes of compounds involved. For example, if the Hamamelidae are characterized by primitive phenolics that are either absent or only moderately represented in the other subclasses, then the Hamamelidae must be a more primitive group (at least with respect to certain secondary compounds) and the others advanced, generally showing a decrease or loss in synthesis of these compounds (cf. Kubitzki & Gottlieb, 1984a, 1984b). Also, the older surveys portray the Magnoliidae and Hamamelidae as being primitive in their flavonoid chemistry due to the overwhelming reporting of flavonols. Yet recent studies on the Winteraceae (Williams & Harvey, 1982), Idiospermaceae (Sterner & Young, 1980), and the Eupomatiaceae (Young, 1983) show not only the presence of flavones in these taxa but also a number of methylated flavones, both advanced characters. If, in fact, flavones and methylated flavones are common in the Magnoliidae, this, along with the absence of the primitive myricetin and ellagic acid (or nearly so), actually suggests a less primitive taxonomic position for the Magnoliidae, and that the more primitive Hamamelidae are not a derivative of the Magnoliidae (sensu Cronquist). Closest chemical similarities of the Hamamelidae lie with some Rosidae, Dilleniidae, and a few Asteridae that have similar compounds, but again, in decreasing amounts (evolution by loss) indicating a more direct relationship with these taxa, or at least parallel biosynthetic capability (Kubitzki & Gottlieb, 1984a, 1984b). To put it simply, one cannot delineate a group (Hamamelidae) as having primitive chemistry and then have it evolve from a group that is possibly more advanced (Magnoliidae) in its chemistry. Alternatively, the possibility still exists that early in their evolution the Hamamelidae and the Magnoliidae may have been more similar in concentrating on synthesis of phenolics. Subsequently, the Magnoliidae and Hamamelidae may have diverged, with the latter retaining emphasis on more primitive phenolics, and the former emphasizing alkaloid synthesis and advanced types of flavonoid substitutions. This, however, is a larger hypothetical "if" (and not provable) than the former alternative, which like all current studies at least deals with factual, contemporary, comparative data. Even in the alternative case, common divergence, rather than derivation, is the logical conclusion to be drawn. Indeed, Kubitzki and Gottlieb (1984a) suggest that the neolignans of the Lauraceae and reduced virolane flavonoid types of the Myristicaceae may represent remnants of an earlier protoangiosperm emphasis on shikimic acid (phenolic) synthesis in these members of the Magnoliidae, where benzylisoquinoline alkaloids are generally absent.

Other compounds (Table 7) such as glucosinolates, sesquiterpene lactones, and polyacetylenes are either limited or erratic in distribution among the angiosperms, providing limited general clarification in angiosperm systematics. However, if we examine biosynthetic and distributional aspects of the nitrogen-containing compounds among the angiosperm subclasses, several interesting comments can be made.

Much has been made of the benzylisoquinoline alkaloids as being characteristic of the Magnoliidae. These compounds (Fig. 6) are derived from aromatic amino acid synthesis. Related isoquinoline and similar types are found in many of the monocots, and apparently in the Caryophyllidae as well. Indeed, if one considers the tyrosine-derived betalains simply as colored alkaloids (Mabry, 1977), then these three taxonomic subclasses are closely related in their biosynthetic origin for these compounds. The Hamamelidae lack these tyrosine-derived alkaloids (at least by present surveys) and thus appear to be less than a direct offshoot of the Magnoliidae. Instead, the Hamamelidae produce mostly nonaromatic amino acid derived alkaloids (Fig. 4) emanating from the citric acid cycle (TCA) or

TABLE 7. Summary of secondary chemistry of angiosperms.

| Taxon | Compounds[a] | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | EA | V | FL | FO | OM | GF | BF | G | CGN | AK | I | S | P |
| Liliidae | − | (+) | Q,K | + | + | + | + | − | O (Ac/k) | O (Ac/k) | − | − | − |
| Magnoliidae | − | (+) | Q,K (M) | + | + | + | (+) | − | O (Ac/k) | O | − | + | + |
| Caryophyllidae | − | (+) | Q,K | + | + | + | − | (+) | O | O | − | (+) | − |
| Hamamelidae | + | + | M,Q,K | (+) | + | + | + | − | Ac/k | Ac/k | + | − | − |
| Dilleniidae | + | + | M,Q,K | + | + | + | + | + | Ac/k | Ac/k | + | + | − |
| Rosidae | + | + | M,Q,K | + | + | + | + | + | Ac/k<br>O | Ac/k<br>O | + | + | + |
| Asteridae | (+) | (+) | Q,K (M) | + | + | + | + | − | Ac/k (O) | Ac/k | + | + | + |

[a] Abbreviations: + = present, − = absent or unknown, (+) = isolated occurrence, EA = ellagic acid, V = *vic*-hydroxylation (= trihydroxylated phenolic ring), FL = flavonols, M = myricetin, Q = quercetin, K = kaempferol, FO = flavones, OM = O-methyl flavonoids, GF = glycoflavones, BF = biflavonyls, G = glucosinolates, CGN = cyanogens, AK = alkaloids, O = aromatic-tyrosine or phenylalanine precursor, Ac/k = non-aromatic amino acid precursor from acetate or Krebs (TCA) cycle, I = iridoids, S = sesquiterpene lactones, P = polyacetylenes.




FIGURE 6. Alkaloid types in the Magnoliidae.

carbohydrate precursors. This is much more like the Rosidae, Dilleniidae, and Asteridae, in which tyrosine-derived alkaloids are absent or begin to decrease in representation in favor of TCA-derived alkaloid precursors, or more importantly in some other groups, toward exploitation of the terpene-steroid-derived pseudoalkaloids. Also, of course, not all Magnoliidae produce isoquinoline alkaloids either.

Consideration of cyanogenic glycosides, however, does suggest a relationship between Hamamelidae and Magnoliidae. Those aromatic cyanogens derived from tyrosine (Fig. 7) are found in the Hamamelidae, Magnoliidae, and Liliidae (Liliopsida) as well as in some Rosidae and Asteridae (Saupe, 1981). Oddly enough, those cyanogens derived from phenylalanine occur in ferns and predominate in advanced angiosperm groups, the Rosidae and Asteridae. The gymnosperms produce tyrosine derived cyanogens like the Magnoliidae-Hamamelidae-Liliidae (Liliopsida). Also non-aromatic cyanogens (valine-leucine, isoleucine) begin to predominate in the Rosidae, Dilleniidae, and Asteridae, and thus may represent the more advanced forms restricted to advanced taxonomic groups, a trend not unlike that observed in the evolution of alkaloids from aromatic to non-aromatic precursors. At least in the cyanogens a more direct relationship between Hamamelidae and Magnoliidae is suggested, although it would be interesting to see if the Ham-

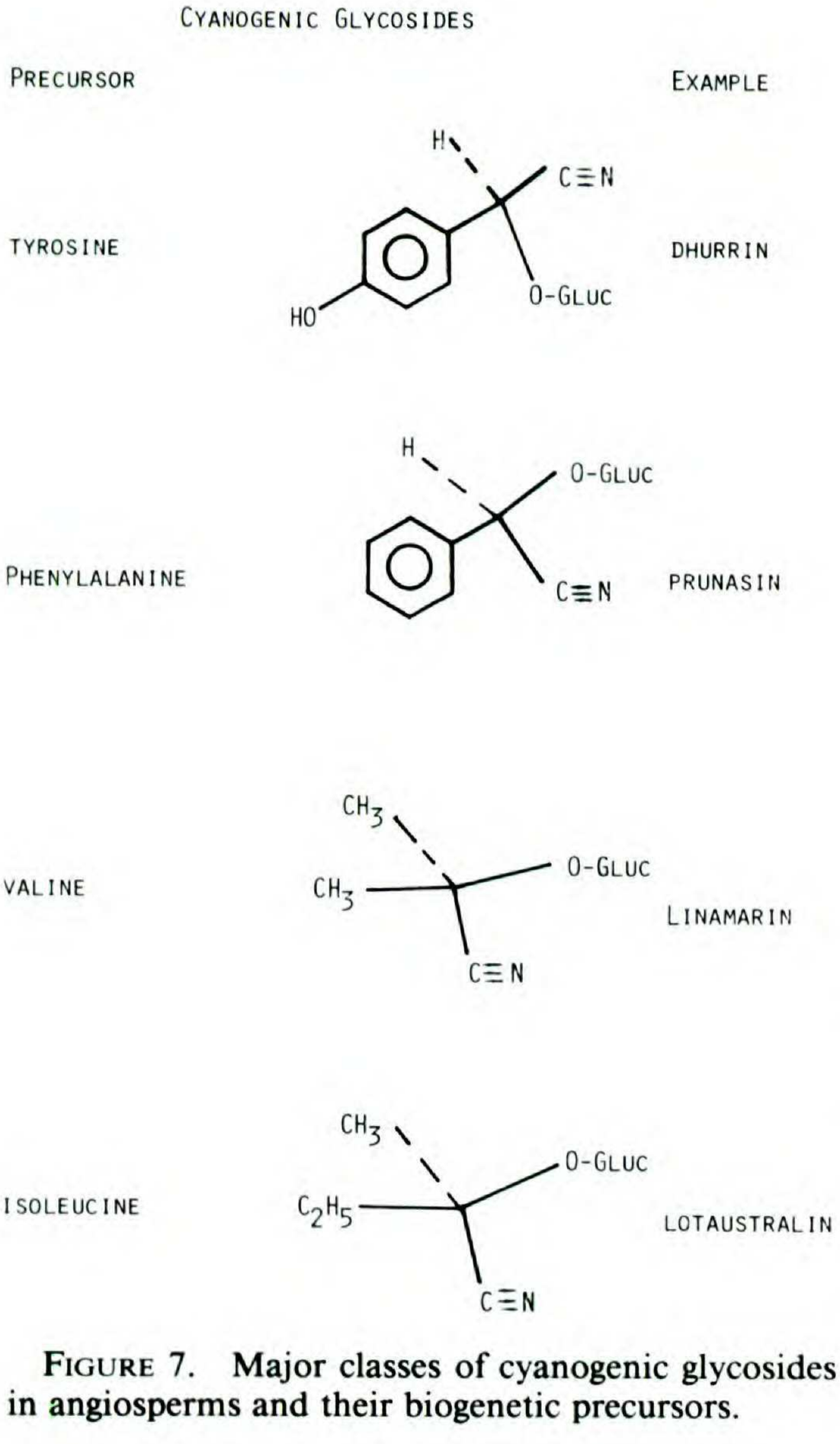



FIGURE 7. Major classes of cyanogenic glycosides in angiosperms and their biogenetic precursors.

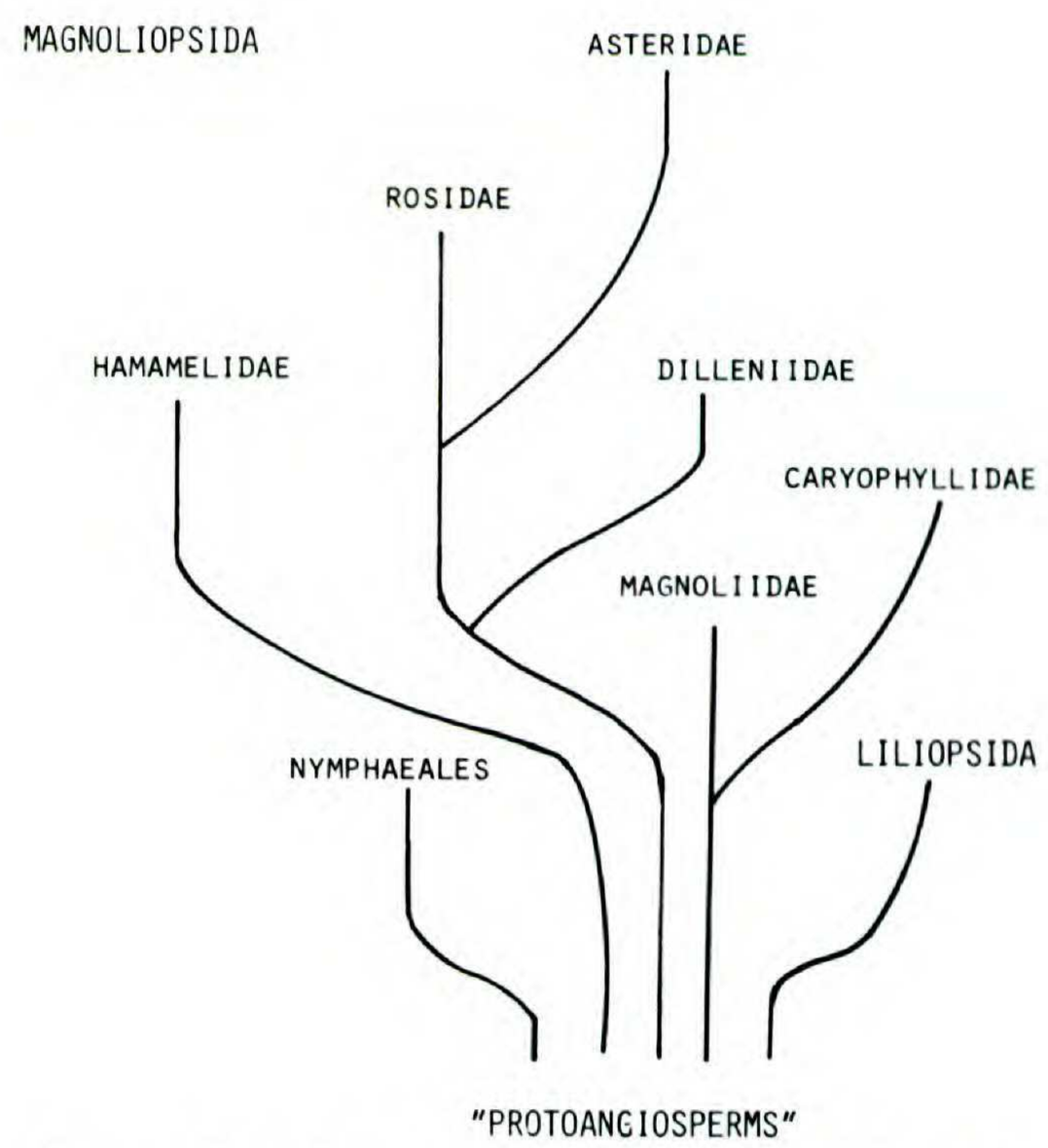



FIGURE 8. Phylogenetic relationship of the Hamamelidae to other subclasses (sensu Cronquist, 1981) based on micromolecular data.

amelidae contain more than one type of aromatic cyanogen and perhaps both aromatic and non-aromatic types. However, among angiosperms there is an overlap in cyanogen types between subclasses (and non-angiosperm seed plants), depending, of course, on the placement of certain taxa within one of the current angiosperm classifications. Thus, some caution should be exercised in emphasizing the systematic significance of these compounds.

Finally, in terms of iridoids, these monoterpene lactone glycosides occur in all of Cronquist's subclasses except Magnoliidae, Caryophyllidae, and Liliidae (Liliopsida). In this case the Hamamelidae are again isolated from the Magnoliidae with suggested similarities closer to the Rosidae.

Looking at Table 7 again, and considering the limitations in conclusions to be drawn from available phytochemical data, I would make the following statements: (1) the Hamamelidae appear to be a primitive subclass of plants at least as old as the Magnoliidae if not older and perhaps not as directly related to the Magnoliidae as suggested, (2) the presence of a number of compounds suggest some similarity (affinity) with the Rosidae/Asteridae, at least at a primal level (this is not new, considering the free movement of taxa by taxonomists between the Hamamelidae/Magnoliidae ↔ Rosidae), (3) the number of unique and highly modified compounds found in some Hamamelidae reinforce the notion of an early divergence of the Hamamelidae from the other subclasses and (4) the current concept of angiosperm monophylesis in the simple sense of a single botanical "Noah's Ark" may require a slightly larger boat or a small but closely integrated fleet, that is, a broader concept of the angiosperm ancestral pool rather than limitation to the Magnoliidae alone (cf. Dilcher, 1979; Retallack & Dilcher, 1981).

The data would suggest an arrangement like that shown in Figure 8, a conclusion similar, at least in part, to the cladistic analysis of non-chemical data for these subclasses by Nixon (unpubl. data), and to conclusions drawn in earlier studies (e.g., Hegnauer, 1977). More recent publications have also come to similar conclusions when attempting to put micromolecular data within biosynthetic and distributional frameworks, recent papers by Kubitzki and Gottlieb (1984a, 1984b), being the most thoughtful and provocative discussions of the problem.

A considerable obstacle to the acceptance of

this argument is the conflict between phytochemistry and palynology. Dilcher (1979) suggests that the reduced, anemophilous, unisexual flower types on catkin-like inflorescences that are found in some possible angiosperm-like Cretaceous fossils represent an alternative ancestral type for modern Hamamelids. However, although the pollen in these fossils may have been wind borne they are monosulcate. Thus they are similar to the earliest presumably angiosperm monosulcate pollen types as found in the Magnoliidae. The presumably more advanced triaperturate pollen of the Hamamelids does not occur as early in the fossil record. Based on pollen distributions then, Hamamelidae still are currently considered to be a derived group (from the Magnoliidae). The problem of opposition in two essentially single morphological character approaches (as well as micromolecules versus pollen) seems insoluble at this point. However, just as phytochemical conclusions may change with each survey, so too each new palynological find may alter current concepts.

The chemotaxonomic debate is by no means finished either, as evidenced by the recent serological review of the angiosperms by Jensen and Greven (1984). These authors indicate that their serological results support a conservative monophylesis of the angiosperms (including the Hamamelidae) from a Magnoliid ancestral group. Interestingly, these serological data also suggest that the Betulaceae are a discordant taxon within the Fagaceae (as do flavonoids), showing a greater similarity to the Magnoliidae. Certainly the rather complex flavonoid chemistry of the Betulaceae (Table 6) does set the family apart within the Fagaceae and the Hamamelidae as well. The same discordance may be cited for the Moraceae (and perhaps the Urticaceae; cf. Table 6), whose flavonoid chemistry is quite unusual within the Urticales and the Hamamelidae generally. Serological work by Petersen and Fairbrothers (1985), in fact, suggests that the Moraceae, Cannabinaceae, and perhaps Urticaceae as well, do not fit in the Hamamelidae, but are better placed near or in the Malviflorae (sensu Dahlgren et al., 1981). Certainly the isoprenyl flavonoids in the Moraceae are found elsewhere only in taxa of the Rosidae/Dilleniidae (sensu Cronquist) lines.

Jensen and Greven (1984) discussed interrelationships among other subclasses as well, and indicated that an expanded survey is desirable (only three taxa tested from the Hamamelidae). Further, they recognized the possibility of changes in their interpretation with the inclusion of more taxa in the test sample. Some considerable variation in precipitation reactions was observed among species from the same genus (*Passiflora*) with the *Magnolia* seed protein employed. Similar exceptional reactions were also observed in some families (Solanaceae). These exceptions and the use of a single seed storage protein still argue for caution in the interpretation of these data.

It is clear from both micro- and macromolecular studies that the Hamamelidae (sensu Cronquist) probably still do not represent a totally natural and homogeneous taxon, and both micromolecular and macromolecular data have much to contribute towards the taxonomic refinement of the concept of the Hamamelidae.

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## APPENDIX I

Voucher specimens used in chemical studies.

*Trochodendron aralioides* Sieb. & Zucc.
UGA Botanical Garden, cultivated
*Tetracentron sinense* Oliv.
*Fang 2725, 6705* (NY)
*Cercidiphyllum japonicum* Sieb. & Zucc.
*Murata et al. 37168, Wood & Boufford 3929* (GA)
*Euptelea pleiospermum* Hook. f. & Thomas
Brooklyn Botanic Garden, cultivated
*E. polyandra* Sieb. & Zucc.
*Boufford 22245, Murata 44430* (GA)
*Platanus acerifolia* Willd.
UGA Campus, cultivated, *Shugrue 55* (GA)
*P. occidentalis* L.
UGA Campus, cultivated
*Corylopsis spicata* Sieb. & Zucc.
Brooklyn Botanic Garden
*C. sinensis* Hemsl.
Brooklyn Botanic Garden, cultivated
*C. pauciflora* Sieb. & Zucc.
Brooklyn Botanic Garden, cultivated
*Distylium lepidotum* Nakai
*Murata et al. 320* (GA)
*D. racemosum* Sieb. & Zucc.
Brooklyn Botanic Garden, cultivated
*Fortunearia sinensis* Rehd. & E. H. Wils.
Brooklyn Botanic Garden, cultivated
*Fothergilla major* Lodd
*Radford 34675, Stewart 1554, Wilbur 7012* (GA)
*F. gardenii* Murr.
*Duncan 5115* (GA)
*Hamamelis vernalis* Sarg.
*Chase 9928* (GA)
*H. virginiana* L.
*Faircloth 4235* (GA)
*H. macrophylla* Pursh.
*Ewan 21059* (GA)
*Liquidambar styraciflua* L.
UGA Botanical Garden
*L. formosana* Hance
UGA Campus, cultivated
*Loropetalum chinense* (R. Br.) Oliv.
*Meyer 16441, Wigginton s.n. 24-XI-51, Coile 2139* (GA)

APPENDIX I. Continued.

*Sycopsis sinensis* D. Oliver.
Brooklyn Botanic Garden, cultivated
*Parrotia persica* C. A. Mey.
Brooklyn Botanic Garden, cultivated
*Sinowilsonia henryii* Hemsl.
Brooklyn Botanic Garden, cultivated
*Myrothamnus flabellifolia* Welw.
*Brass 16132, Cronquist & de Winter 11608* (NY)
*Daphniphyllum teijmanensis*
Brooklyn Botanic Garden, cultivated
*D. glaucescens* Blume
Brooklyn Botanic Garden, *Tanaka & Shimada 17827*
*D. calycinum* Benth.
Brooklyn Botanic Garden, *Levine 1624*
*Eucommia ulmoides* Oliv.
Brooklyn Botanic Garden, cultivated; *Gillis 14345* (GA)
*Humulus americanus* Nutt.
*Chase 12151* (GA)
*H. japonicus* Sieb. & Zucc.
*Windler & Stastny 4046* (GA)
*Broussonetia papyrifera* L'Her
GA, Clarke Co., cultivated
*Cudrania tricuspidata* (Carr.) Bur. ex Lavallee
GA, *Deason s.n. XI-81,* cultivated
*Dorstenia foetida* Schweinf. (= *D. obovata* Hochst.)
UGA Greenhouses, cultivated
*Fatoua villosa* (Thunb.) Nakai
*Godfrey 72357, Thieret 10227* (GA)
*Ficus aurea* Nutt.
*Stinson 240* (GA)
*F. benjamina* L.
*Brumbach 9711* (GA)
*F. brevifolia* Nutt.
*Scull s.n. 27-I-40* (GA)
*F. caprifolia* Del.
*Russel 2054* (GA)
*F. carica* L.
*Rainwater E8113* (GA)
*F. citrifolia* Mill.
*Brumbach 9770* (GA)
*F. gemina* Ruiz ex Miq. in Mart.
*Rimachi 2790* (GA)
*F. laevigata* Vahl.
*Smith 966* (GA)
*F. llewellynii* Standl.
*Rimachi 1778* (GA)
*F. macrophylla* Desp.
*Stimson 2050,* cultivated (GA)
*F. nitidifolia* Bur.
*Bauman-Bodenheim 15156* (GA)
*F. perforata* L.
*Sauleda 3733* (GA)
*F. pumila* L.
*Crawford et al. 959* (GA)
*F. webbiana* Miq.
*Guillaumin 9178* (GA)

APPENDIX I. Continued.

*Helicostylis elegans* (Macbr.) C. C. Berg.
*Rimachi 4029* (GA)
*H. scabra* (Macbr.) C. C. Berg.
*Rimachi 2898* (GA)
*Morus alba* L.
*Faircloth 5308, Redfearn 3712,* Clarke Co., cultivated (GA)
*M. rubra* L.
*Duncan 5131,* Clarke Co., cultivated
*Cecropia peltata* L.
*Duke 12500* (GA)
*Pourouma palmata* P. & E.
*Rimachi 2724* (GA)
*P. phaeotricha* Mildbr.
*Rimachi 2725* (GA)
*Boehmeria cylindrica* (L.) Sw.
*Adams & Duncan 19488, Hardin 14286* (GA)
*Laportea canadensis* (L.) Wedd
*Pease s.n. 30-VII-58, Pyron & McVaugh 857* (GA)
*Urtica chamaedryoides* Pursh
*Thieret 32699* (GA)
*U. dioica* L.
*Clokey 8322, Swendsen 487* (GA)
*Leitneria floridana* Chapm.
*Demaree 45267E, McDaniel 903; E. L. Richards 9749* (STAR)
*Rhoiptelea chiliantha* Diels & Handel-Mazzetti
*Ching 5840* (NY)
*Myrica asplenifolia* L. (= *Comptonia perigrina*)
*Ahles 75334, Hardin 364, Hunt MA 180* (GA)
*M. cerifera* L.
*Faircloth 3444, Lane 142* (GA)
*M. gale* L.
*Miller E4315, Ahles 89166* (GA)
*M. heterophylla* Raf.
*Hardin & Duncan 14573, 20724* (GA)
*M. inodora* Bartr.
*Faircloth 732, Godfrey & Harrison s.n. 9-III-57* (GA)
*M. rubra*
*Wigginton s.n. 24-XI-51, s.n. 30-XII-52* (GA)
*M. serrata* Lam.
*Russel 2093* (GA)
*Casuarina equisetifolia* Forst.
*Dugger s.n. 29-IX-40, Ward & Crosby s.n. 9-VIII-65* (GA)
*C. glauca* Sieb. ex Spreng.
*Baum & Wilson 161* (GA)
*C. cunninghamiana* Miq.
*Duncan 30462* (GA)